

MEASUREMENTS OF (N, XN) CROSS SECTIONS FOR HYBRID SYSTEMS

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Abstract

In this paper we will report on tests we have performed at GELINA (IRMM Geel), CYCLONE (Louvain-la-Neuve) and n-ToF (CERN) to demonstrate that the in-beam γ -ray spectroscopy can be adapted to low frequency white neutron beams or to quasi-monoenergetic neutron beams in order to measure (n,xn) cross sections. After a description of the nuclear data status, regarding the hybrid system thematic, of the (n,xn) reactions and of the in-beam γ -ray techniques, the results on $^{207}\text{Pb}(n,2n)$, $^{206,207,208}\text{Pb}(n,n')$ and $^{232}\text{Th}(n,5n)$ preliminary measurements will be presented. Real measurements of $^{207}\text{Pb}(n,2n)$, $^{207}\text{Pb}(n,n')$ and $^{232}\text{Th}(n,5n)$ have been performed from April to July 2004, preliminary results will be discussed.

Introduction

Nuclear data status

The design of a hybrid reactor (ADS) and even that of a demonstrator, as well as the study of the thorium cycle potentialities require the knowledge of a wide range of better nuclear data. Today the data bases are relatively well filled for neutron energies below 20MeV and for nuclei involved in the Uranium cycle, but for those involved in the Thorium cycle and for high neutron energies the situation is not satisfactory. The experimental data are very scarce and if they exist, are too imprecise. Consequently real discrepancies exist between evaluated data, and the data bases are often fed by model predictions. This situation is illustrated by a simulation [1] which studies the discrepancies obtained on the reaction rate in a hybrid system for calculations performed with different data bases as input file and for three types of reaction. For fission and capture of actinide nuclei, the variation is generally fairly small. For some nuclei it may reach however 10% for fission and 25% for capture. For the (n, xn) reactions, the average discrepancy is around 20% and reaches 350% for ^{238}Pu . The situation is quite similar for nuclei representative for the spallation source or for the structure materials, since the average discrepancies are about 10-15%. Another calculation [2] has shown that with the present error on (n, n') and (n, 2n) cross sections on lead, the precision on the criticality of an ADS is not better than 2 %. This is, indeed, not acceptable.

A few years ago the scientific community realised that there was a real and urgent need of new and more precise data for nuclei representative of the core, of the spallation target and of the structure materials of a hybrid system. These measurements should contribute to better evaluated data, but since all reactions cannot be measured they should also provide adequate constraints on nuclear reaction models like the intra-nuclear cascade or the preequilibrium model. Consequently they would improve their predictive power and the quality of Monte Carlo simulation codes like FLUKA or MCNP.

(n,xn) reactions

The (n,xn) reactions which do not play a crucial role in classical reactors except for x=1 because of their high energy threshold, which increases with increasing x value, become more important in fast reactors and even more in hybrid systems. Indeed they participate to the neutron multiplication in the spallation target (Pb/Bi). Fast neutrons can also escape from this target, reach the core and then interact with fuel materials by (n,xn) reactions. Their role is also important if we consider the thorium cycle. Indeed the $^{232}\text{Th}(n,2n)$ reaction leads to ^{231}Th which decays to ^{231}Pa that can dominate the radiotoxicity in some scenarios of fuel reprocessing. [2] Another example concerns the $^{233}\text{U}(n,2n)$ reaction which leads to ^{232}U and which decays, after several α and β emissions, on the excited states in ^{208}Pb . This last nucleus decays by emitting a high energy (2.6 MeV) γ ray that makes the fuel elaboration more complicated.

The data concerning (n,xn) reactions are scarce because of the difficulty to measure their cross section. Three techniques exist with their own characteristics and limitations. The more often used is the activation method. It requires that the final nucleus is unstable and has a half life suitable for activity counting, usually in the range between a few seconds and several years. A second method is the direct measurement of the emitted neutrons. The strong constraint of this method is the efficiency of the detector which has to be theoretically of 100% to distinguish the different (n,xn) reactions. As this is not possible corrections have to be applied which limit the precision. This limitation is even stronger if the sample is fissionable. In this case it is difficult to separate neutrons from fission from those from (n,xn) reactions. These two methods require a monoenergetic beam. The third technique, the in-beam γ -ray spectroscopy, has been strongly developed in heavy ion induced reactions physics. One advantage of this method is that it is the possibility to use it also on white neutron beams. Nevertheless difficulties have to be overcome. One of them is related to γ -detection performed with Germanium detectors (see the following sub-section) which are very sensitive to neutron radiation. Therefore, the beam has to be very well collimated to avoid damaging of the detector crystal.

The in-beam γ -ray spectroscopy for (n,xn) reaction studies

The in-beam γ -ray spectroscopy takes advantage of the well known nuclear level schemes for nuclei near the stability. The (n,xn) reaction leaves generally the residual nucleus in an excited state. The deexcitation implies the emission of γ -rays which allow identifying it unambiguously if the energy resolution is sufficient, and if large parts of the γ -cascades pass through the first levels which then decay to the ground state. Because of their high energy resolution the High Purity Germanium (HPGe) detectors are used for this type of measurements.

At a white neutron beam, time of flight measurement is applicable to determine the neutron energy if the flight path is sufficiently large. For example a 1 MeV resolution on the neutron energy is achieved up to 20 MeV with a time resolution of 10 ns and a flight path larger than 30 m.

By this method we can extract directly from the measurement the (n,xn γ) cross section corresponding to a specific transition. To obtain the total (n,xn) cross sections, corrections have to be applied, like side feeding or corrections related to the presence of isomeric states (see next section). This information is given by theoretical models. The correction depends strongly on the studied nucleus. If the first transition is measured, like it is the case for our $^{207}\text{Pb}(n,2n)$ measurement, the side feeding correction is of the order of 10-15%.

(n,xn) cross section measurements

Measurements at Cyclotron CYCLONE - Louvain-la-Neuve

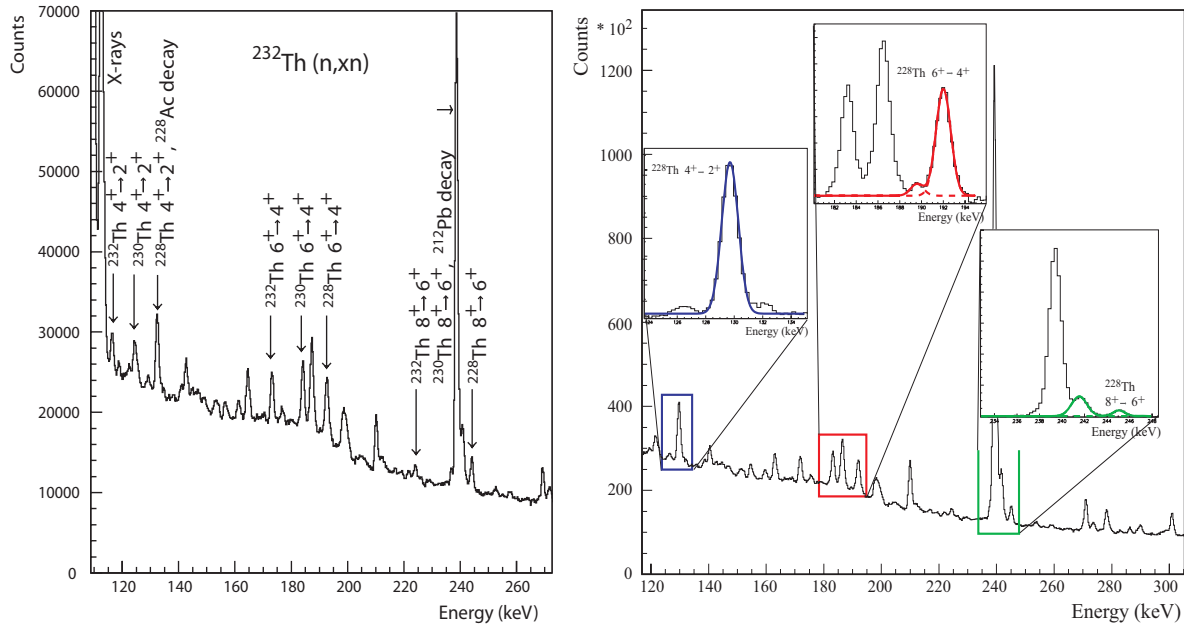
Neutron beam

The neutron beam is produced by the interaction of a proton beam delivered by the CYCLONE [4] cyclotron (proton energy available from 25 up to 70 MeV and proton current about 10 μA) with a Lithium target (3 or 5 mm thick) thanks to the $^7\text{Li}(p,n)^7\text{Be}$ reaction (Figure). A typical neutron energy spectrum consists of a well defined peak (about of 3 MeV resolution) which contains, for example at 65 MeV proton energy, about 50% of the neutrons and a nearly flat low energy continuum.

Experimental set-up and results of the tests

For tests [5] with a ^{232}Th sample (0.3 mm) we have placed a Thorium sample at 3.3 m from the neutron source and a 1.5 cm deep HPGe Planar detector at 12 cm from the sample. The acquisition time was about 17 hours with a 38 MeV neutron energy beam. The results are shown in Figure where we can see several lines corresponding to the $4^+ \rightarrow 2^+$, $6^+ \rightarrow 4^+$ and $8^+ \rightarrow 6^+$ transitions in ^{232}Th fed by the $^{232}\text{Th}(n,n')$ reaction, in ^{230}Th fed by the $^{232}\text{Th}(n,3n)$ reaction and in ^{228}Th fed by the $^{232}\text{Th}(n,5n)$ reaction.

Figure 1. Portion of prompt γ spectrum (left) obtained in 17 hours with a ^{232}Th sample at 38 MeV incident neutron energy peak and zoom on the interesting peaks (right)

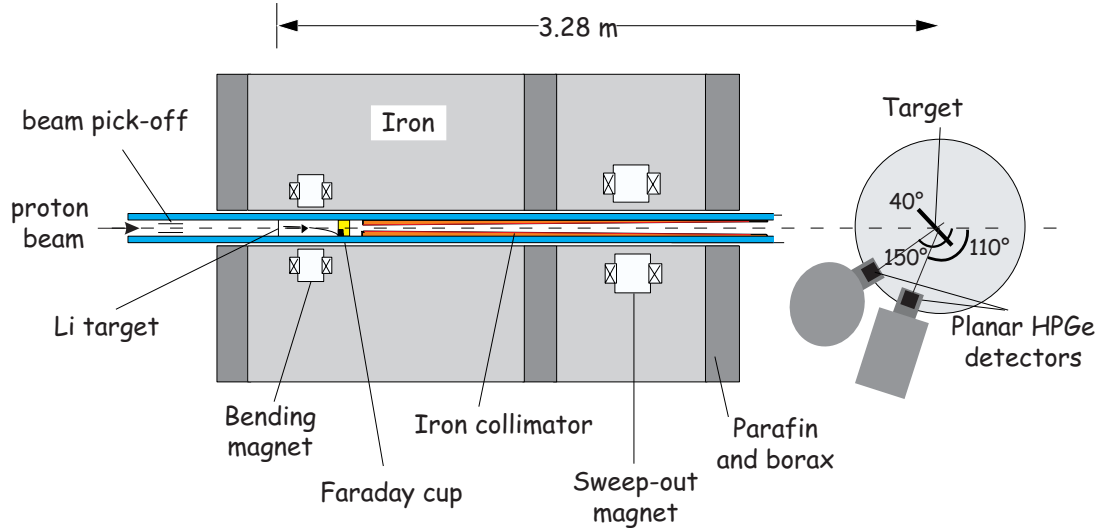


This short test has proved that we can extract the peaks corresponding to the interesting γ transitions in three Th isotopes and that it will be possible to extract the excitation function of the $^{232}\text{Th}(n,5n \gamma)$ for three cascading γ transitions.

$^{232}\text{Th}(n,5n \gamma)$ measurements

After the good results of the tests, the real experiment has been performed this summer (July 2004) at Louvain-la-Neuve. For this experiment 27 shifts (each of 8 hours) were allocated during which we have managed to do measurements at six neutron energies around 38 MeV (the estimated maximum of the cross section) to cover the excitation function energy range. Two germanium detectors were used for γ detection (see Figure 2).

Figure 2. **Global view of the experimental set-up of the neutron beam and detectors used for detection.**



Indeed with two detection angles, we can minimize the angular distribution effect in the determination of the angle integrated cross section [6, 7]. As all the interesting transition are pure E2 transitions the angular distribution can be describe in terms of Legendre polynomials. The Gauss quadrature specialised to Legendre polynomials allows then to write that

$$\sigma = \int \frac{d\sigma}{d\Omega}(\vartheta) d\Omega = 2\pi \left(w_1 \frac{d\sigma}{d\Omega}(\vartheta_1) + w_2 \frac{d\sigma}{d\Omega}(\vartheta_2) \right)$$

where

$$w_1 = 0.695 \rightarrow \vartheta_1 = 150^\circ, 30^\circ$$

$$w_2 = 1.305 \rightarrow \vartheta_2 = 70^\circ, 110^\circ$$

With two detectors placed at those angles, only the contribution of Legendre polynomials of order 8 or higher to the angle integrated cross section is neglected.

As said previously, the neutron beam is not monoenergetic and there is a non negligible continuum part of neutrons. In this case, the values of the cross sections follow from the solution of the integral equations

$$\int \sigma(n,5n\gamma; E) \cdot \Phi_{nE}^k(E) dE \propto \frac{N_{\gamma}^k}{N_{\text{targ}}}$$

where $k=1,\dots,n$ is a label used to distinguish measurements done with neutron beams of different peak energies and N_{γ}^k is the number of counts in the interesting γ peak for the k -th measurement, Φ_{nE}^k , the neutron spectral fluence for the k -th measurement and N_{targ} is the number of ^{232}Th nuclei in the target. One method to select the monoenergetic part would have been to perform time of flight measurements with the Germanium detector and to select events coming from the peak. Unfortunately, the time resolution of the detector (about 10 ns) is not sufficient, for a flight path of 3 m, to achieve a sufficient neutron energy resolution. Another way to extract this monoenergetic part, and then correct our data, is to measure the fluence as a function of the neutron energy and also the absolute value of the number of neutrons in the peak. With this information we could extract the proportion of neutrons coming from the peak. For example at a proton energy of 40 MeV the continuum part of the neutron spectrum (above the threshold energy of $^{232}\text{Th}(n,5n)$ reaction, i.e. ~ 24 MeV) represents only 35% of the peak component. All these flux measurements were done using instruments and methods developed by the PTB group for the UCL neutron beam facility [8]. The PTB proton recoil telescope PRT2 has been used to measure the neutron fluence of the high-energy peak for each proton energy used. The neutron energy distribution has been measured with a liquid scintillation detector and the beam monitors (fission ionization chamber, plastic scintillation detector) have been used to correct for possible variations of the neutron yield between the fluence measurement and the $(n,xn\gamma)$ experiment. We plan to perform the deconvolution with the help of standard unfolding methods which are used in the maximum entropy unfolding code MAXED [9]. One of the strengths of this proposed method is its ability to incorporate prior information into the unfolding problem in a systematic and mathematically well defined way. This is of interest here, because some prior information about the cross sections will be available (i.e. from nuclear model calculations). The neutron spectral fluences that are needed for the unfolding will be determined from the above mentioned measurements. These methods permit the use of standard procedures for sensitivity analysis and propagation of uncertainties.

The data analysis is now in progress to extract the cross sections of the $^{232}\text{Th}(n,5n\gamma)$ reaction for the six neutron energies used. To extract the total $(n,5n)$ excitation function theoretical work is also needed to calculate precisely the feeding of the ground state by deexcitation chains bypassing the transitions observed in our experiment.

Measurements at GELINA – Geel

Neutron beam

The GELINA facility [10, 11] is composed of an 800 Hz pulsed electron beam (electron energy available from 70 to 140 MeV) which hits a rotating U wheel. The bremsstrahlung process induces photofission of the uranium nuclei which produces an isotropic neutron flux with a broad energy spectrum. This neutron flux can be used at sixteen different flight paths.

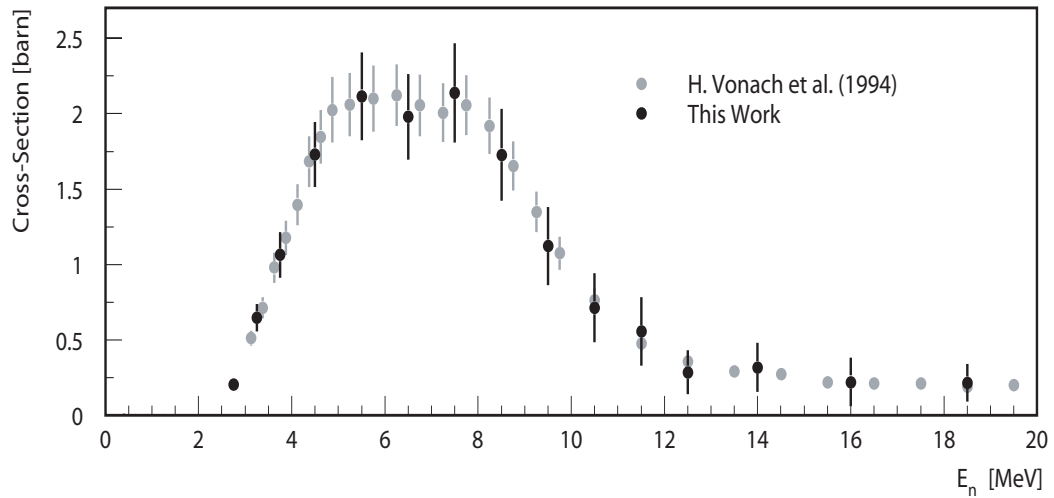
Experimental set-up and results [12] of the tests with a natural Lead sample

The area used for the preliminary measurements [13] on a natural lead sample (6 mm thick) was located at 200 m from the neutron source. An HPGe Clover detector was placed at 12 cm from the sample and the acquisition time was 85 hours. To ensure the possibility to record γ rays coming from high energy neutron reaction despite the presence of a strong γ flash, we have chosen to use a digital acquisition [14]. Indeed with a classical acquisition, the voltage at the output of the amplifier when a γ from the flash has been detected, recovers its baseline after several μ s only. There is therefore an upper limit on the neutron energy above which the γ energy cannot be measured accurately. In our case at GELINA, this limit corresponds to a few MeV which is not sufficient to study the (n,xn) reactions with x greater than 1.

We have used a home made digital acquisition (14 bit, 65Ms/s converters) and developed an analytic treatment of the digitalized signal which reduces the dead time without worsening the energy resolution. With the HPGe Clover detector we have achieved a dead time of about 3.2 μ s which allows measurements up to 14 MeV neutron energy with a γ energy resolution of about 3.1 keV at 2.6 MeV (i.e. for the ^{208}Pb ground state transition). Higher neutron energies can be attained with a slightly worse resolution.

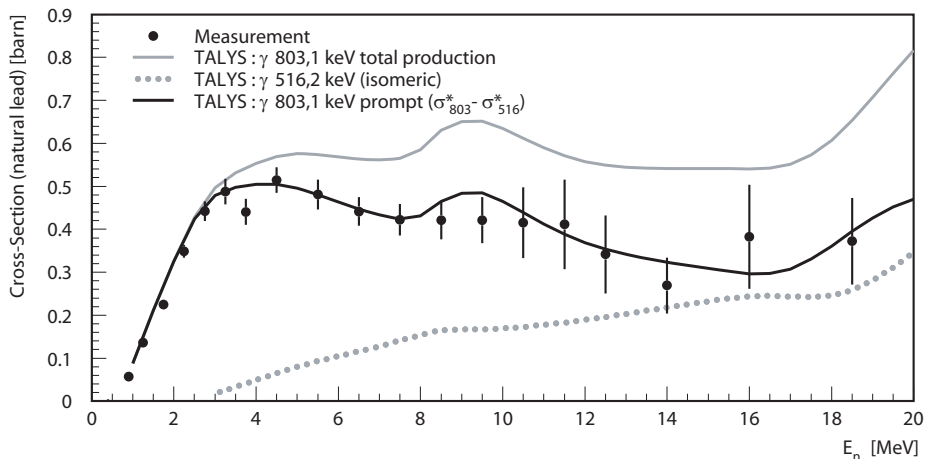
Since the sample was natural lead, several reactions are possible. The excitation functions for the 803.1 keV transition in ^{206}Pb is the sum of the contribution of the $^{206}\text{Pb}(n,n'\gamma)$, the $^{207}\text{Pb}(n,2n\gamma)$ and the $^{208}\text{Pb}(n,3n\gamma)$ reactions. The 569.7 keV transition in ^{207}Pb is the sum of the contributions of the $^{207}\text{Pb}(n,n'\gamma)$ and $^{208}\text{Pb}(n,2n\gamma)$ reactions, while the 2614.5 keV in ^{208}Pb is fed almost only by $^{208}\text{Pb}(n,n')$ (the $^{207}\text{Pb}(n,\gamma)$ cross section is very weak compared to the $^{208}\text{Pb}(n,n')$ cross section). For the tests, the absolute neutron flux and the clover efficiency have not been measured. The absolute cross sections have then been determined by normalizing the measured data to the TALYS [15] predictions [16]. To calculate the normalization factor, the data of the 2614.5 keV transition in ^{208}Pb has been used as it can only be generated by the $^{208}\text{Pb}(n,n')$ reaction. Figure shows the comparison between our work and the one from Vonach [17] et al. for the cross sections of the 2614.5 keV transition in the $^{208}\text{Pb}(n,n')$ reaction.

Figure 3. Cross section of the first transition in the ^{208}Pb obtained by the reaction $^{208}\text{Pb}(n,n')$



To obtain the absolute cross section of the other transitions, the normalization factor is simply deduced by correcting the one found for the 2614.5 keV transition in ^{208}Pb by the efficiency and the self absorption effect. In the ^{206}Pb and ^{207}Pb level schemes, one has to note the presence of an isomeric state (respectively 1.6037 MeV and 516 keV) with a half life of respectively 125 μs and 800 ms which are long compared to the neutron time of flight range. The γ decay of these isomeric states cannot thus be correlated to the neutron burst which has created it. Therefore the measured prompt γ yield represents the difference between the relevant transition yield and its delayed part. The TALYS code allows taking this isomeric state effect into account. Figure 4 shows the different contributions of the first transition in ^{206}Pb .

Figure 4. Different contribution to the production cross section of the first transition in ^{206}Pb given by TALYS compared to our data.



As mentioned previously, this transition is also a sum of different reaction contributions which have been separate using the TALYS code. The results are shown in for the 803.1 keV transition in ^{206}Pb .

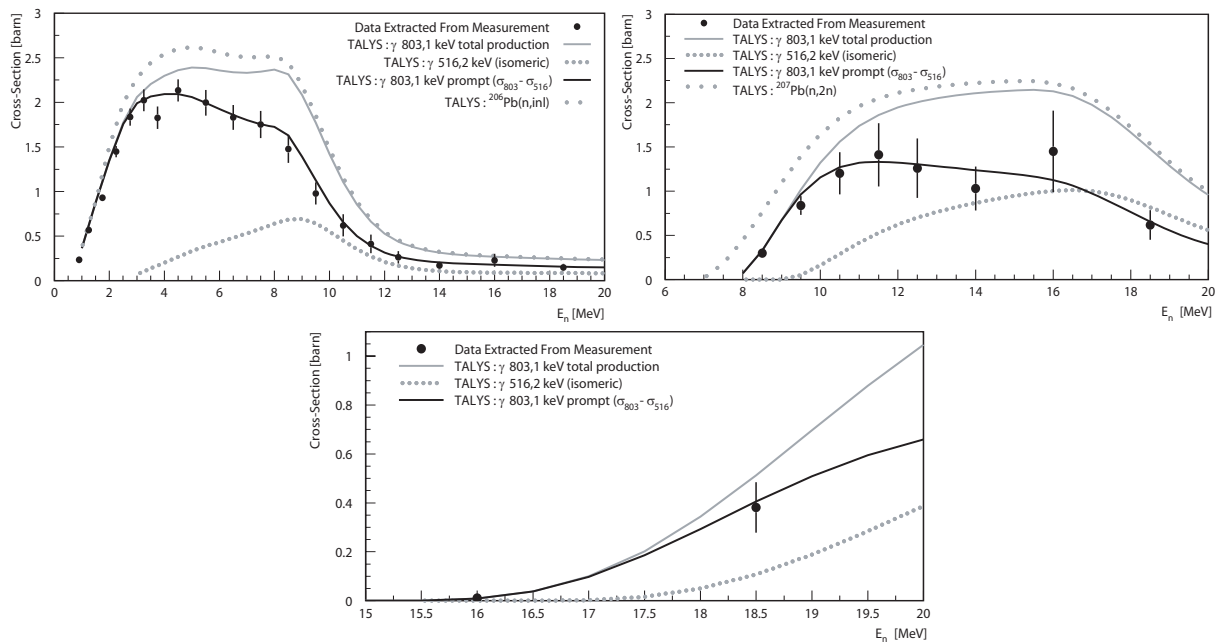
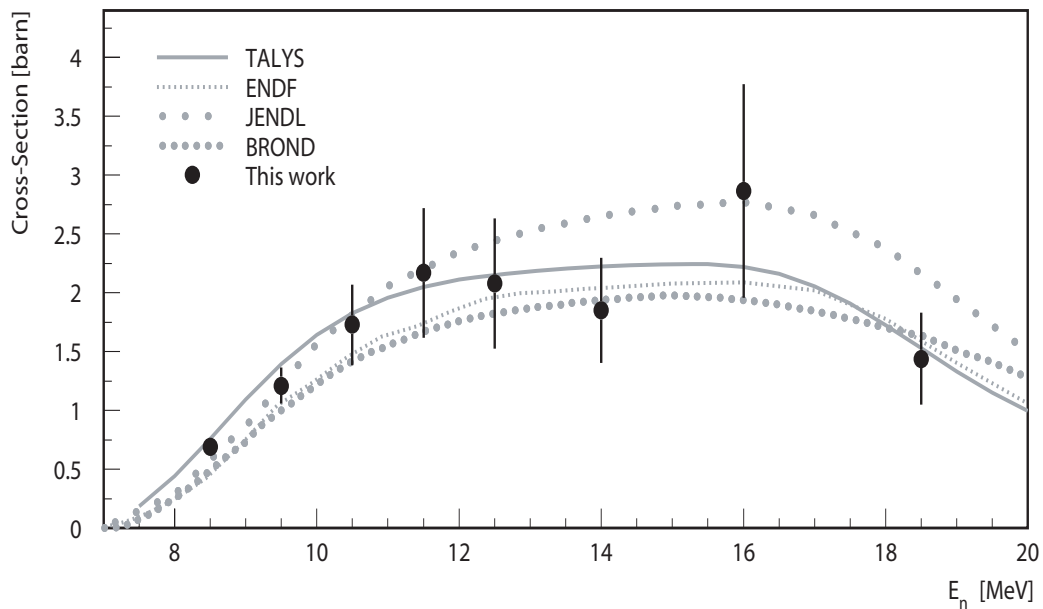


Figure 5. The three contributions ($^{206}\text{Pb}(n,n')$ top-left, $^{207}\text{Pb}(n,2n)$ top-right, $^{208}\text{Pb}(n,3n)$ bottom) of the 803.1 keV transition distinguished with the TALYS calculations.

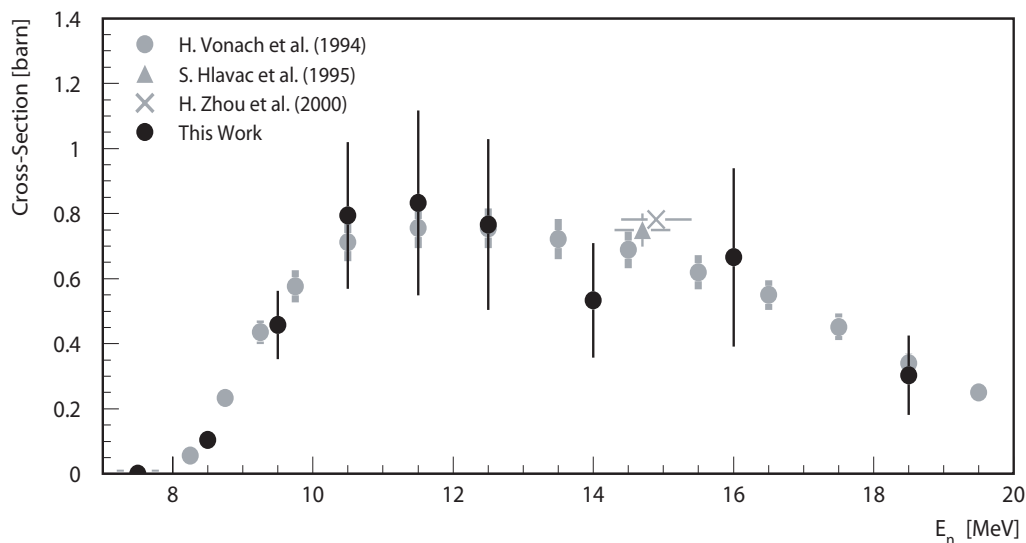
The data obtained for the $^{207}\text{Pb}(n,2n)$ cross section are compared to the data base prediction in Figure 6.

Figure 6. Comparison of our data to different data base evaluations of the $^{207}\text{Pb}(n,2n)$ cross section.



One notes that our data are in good agreement with the data bases at low energies but at high energies the errors are too large (the neutron flux is very low) to allow to conclude on the validity of one base. Nevertheless we can note that the results are fairly in agreement with the theoretical predictions. Finally the results concerning the $^{208}\text{Pb}(n,2n\gamma)$ are presented in Figure 7 where they are compared to previous experimental results. We can note the good agreement between all these experimental data.

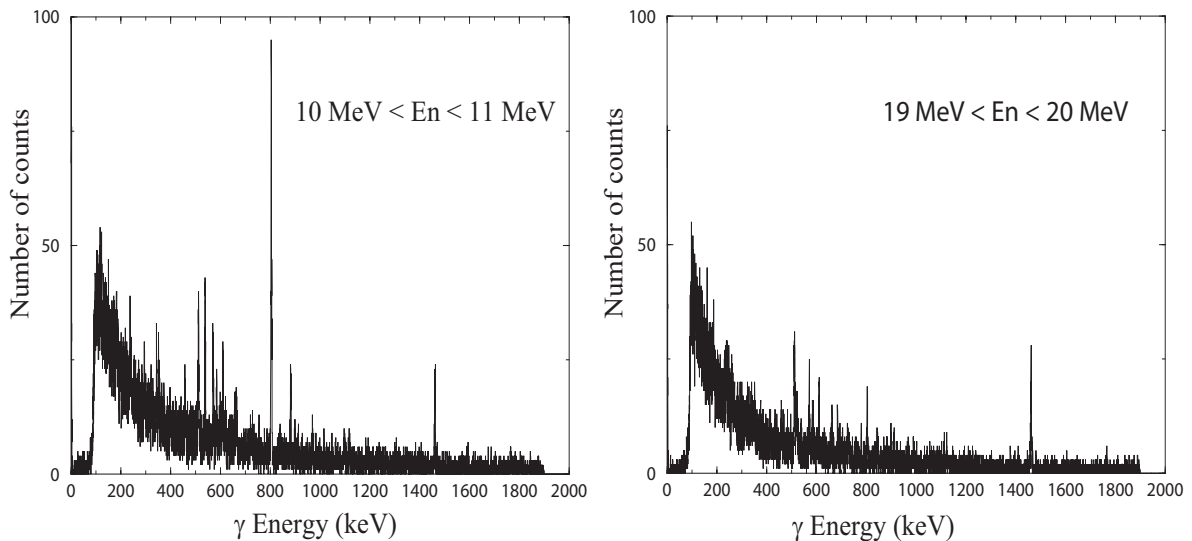
Figure 7. Comparison of our data for the first transition in the $^{207}\text{Pb}(n,2n\gamma)$ reaction to data from Vonach et al. [17] (1994), S.Hlavac et al. [18] (1995), H.Zhou et al. [19] (2000).



Measurements with an enriched ^{207}Pb sample

The experiment was done at GELINA from May to July of this year. Two coaxial germanium detectors have been used and the total acquisition time was in the order of 700 hours. With this time the statistic is good even at high energy as it is shown in the preliminary results (Figure 8) which represents the prompt γ spectrum for low energy neutron bin (10 to 11 MeV, (left)) and high energy neutron bin (19 to 20 MeV, (right)) for the 100% coaxial HPGE detector.

Figure 8. Prompt γ spectra corresponding to the $^{207}\text{Pb} + n$ reaction and for $E_n=10-11$ MeV (left) and for $E_n=19-20$ MeV (right)



The first case corresponds to the neutron energy near the maximum of the $(n, 2n)$ cross section and the spectrum is dominated by the 803.1keV transition. In the second case for which the flux and the cross section are lower the total counts in the 803.1keV peak is still around 300. The analysis of these data is now in progress and we hope to produce sufficient accurate results to test the evaluated prediction.

Tests at n-TOF at CERN

Neutron beam

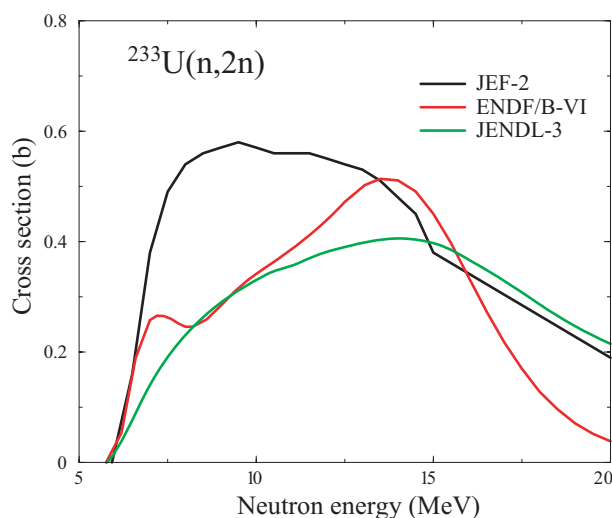
The neutron beam at n-TOF is produced by the interaction of a 24GeV proton beam of the PS with a Pb target of 80x80 cm². The neutrons are created by spallation reactions and their energies cover the large range from 1eV to 100MeV. The PS has a super-cycle of about 14 s and delivers a few bursts which contain typically some $7 \cdot 10^{12}$ protons per burst, within each super-cycle.

Results of tests

Three tests have been done at n-TOF with a planar HPGe detector. For the first one, the background due to muons was too high and blinded all the detectors. After a partial reduction of this background we have noted, during a second test, that the muons were observed only near the beam but a classical preamplifier was still blocked at each burst during more than 100 μ s. Indeed the energy deposited in the crystal is very large since it can reach up to 150MeV. For the third test we have equipped our detector with an external reset preamplifier. This preamplifier was specially designed to inject an inverse current triggered by the accelerator signal to counterbalance the flash. The detector was then ready again after as much as 10 μ s only. This is a clear improvement, but the maximum recovery time is still too high.

The solution is of course to reduce the muon background. This seems possible since it has already been achieved in almost the entire experimental hall, decreasing thereby the neutron background by more than an order of magnitude. The addition of an iron shielding in the vicinity of the beam tube has been done during the last winter shut-down. We plan thus to perform a test during this autumn before the shut down of the beam. If it is successful we will check, in the future the feasibility of the $^{233}\text{U}(n,2n)$ cross section measurement. Though good knowledge of the $^{233}\text{U}(n,2n)$ reaction is crucial in the thorium cycle, as shown in the Figure 9 present data bases present large discrepancies and no experimental data exist.

Figure 9. Comparison between three different data bases for the $^{233}\text{U}(n,2n)$ cross section



Conclusion

We have demonstrated that the in beam γ -ray spectroscopy can be used at the neutron beams GELINA (Geel) and Cyclotron CYCLONE (Louvain-la-Neuve) despite their very different characteristics. Besides a new test in CERN to check the efficiency of the new shielding is planned, we have measured the $^{207}\text{Pb}(n, n')$ and $^{207}\text{Pb}(n,2n)$ up to 20MeV neutron energy with an enriched ^{207}Pb sample at IRMM and the $^{232}\text{Th}(n,5n \gamma)$ excitation function from threshold to 45 MeV at Louvain-la-Neuve. Data analysis of these experiments is in progress. We also plan to measure the (n, n') and $(n,2n)$ cross sections on ^{206}Pb and ^{208}Pb at GELINA. Furthermore, it is our particular interest to proceed with measurements on structural materials in Louvain-la-Neuve.

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